Weak localization of light in superdiffusive random systems

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Lévy flights constitute a broad class of random walks that occur in many fields of research, from animal foraging in biology, to economy to geophysics. The recent advent of Lévy glasses allows to study Lévy flights in controlled way using light waves. This raises several questions about the influence of superdiffusion on optical interference effects like weak and strong localization. Super diffusive structures have the extraordinary property that all points are connected via direct jumps, meaning that finite-size effects become an essential part of the physical problem. Here we report on the experimental observation of weak localization in Lévy glasses and compare results with recently developed optical transport theory in the superdiffusive regime. Experimental results are in good agreement with theory and allow to unveil how light propagates inside a finite-size superdiffusive system.

Lévy flights are maybe the most general class of random walks, of which the commonly known Brownian motion is a limiting case [1, 2]. They are governed by Lévy statistics[3], which have the fascinating property that, depending on the value of one control parameter, they can exhibit a diverging variance [4]. This leads to a phenomenon called superdiffusion, being a diffusive process in which the mean square displacement increases faster than linear in time [5, 6]. Lévy flights are common in nature and appear, for instance, in animal food searches [7, 8], laser cooling of cold atoms [9], evolution of the stock market [10], astronomy [11], random lasers [12] and turbulent flow [13].

The recent development of Lévy glasses [14] and carefully prepared hot atomic vapours [15], have allowed the observation of Lévy flights of light waves and the resulting superdiffusion process. Since interference effects play a dominant role in light transport, this raises the natural question how interference influences optical superdiffusion - a concept which has not been addressed so far. In regular - diffusive - disordered optical systems, interference leads to speckle correlations, and weak and strong localization effects, which all have been studied extensively over the last two decades [16, 17]. Localization, in particular, leads to a complete halt of transport that can confine light waves in random patterns. Since the superdiffusion induced by Lévy statistics tends to enhance transport, one would expect it to counter-act localization induced confinement.

Among all interference phenomena in random optical materials, maybe the most robust is that of weak localization [16]. It is observed in the form of a cone of enhanced backscattering, which contains information on the path length distribution deep inside the random system, and which has been observed in recent years from several diffusive random structures [18–25] Some of us have recently shown theoretically that weak localization, or coherent backscattering, can be observed from Lévy glasses and that a superdiffusive approximation can be used to predict its behaviour [26]. In this paper we report on the experimental observation of weak localization from superdiffusive materials, which constitutes the first observation of an interference effect in transport based on Lévy statistics. We find a good agreement with superdiffusive transport theory and show how the backscattering cone can be used to extract the Green's function in a Lévy glass. Contrary to regular diffusive media, in a Lévy glass, light from an *arbitrary* depth inside the medium has a nonvanishing probability to couple directly to the surrounding environment. This latter property makes light scattering from Lévy glasses complex, and has important consequences for its (back)scattering properties.

The Lévy glasses under investigation are made of jammed-packed microscopic glass spheres, whose diameter (ϕ) varies almost over two order of magnitudes (from 5 to 230 µm) following a power-law distribution $p(\phi) \propto \phi^{-(\beta+1)}$, with β adjustable parameter. These spheres are embedded in a polymeric matrix which matches their refractive index ($n = 1.52$) and in which TiO_2 nanoparticles (average diameter 280 nm) have been dispersed (see Fig. 1(a)) [27]. Because of their refractive index ($n = 2.4$) higher than the polymer, these nanoparticles act as point scatterers which are not homogenously distributed throughout the sample due to the presence of the glass spheres (see Fig. 1(b)). As a result, light transport is dominated by the long "jumps" that light performs propagating through the microscopic spheres. The step length distribution that light performs in Lévy glasses fol-
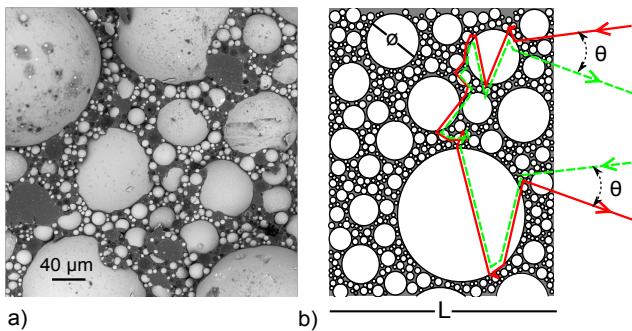
FIG. 1: (a) Electron micrograph of the interior of a Lévy glass. (b) Sketch representing the optical mechanism laying behind the coherent backscattering cone in a Levy glass. L=300 µm is the thickness of the sample and ø the diameter of the spheres.

lows a power-law decay as $p(l) \propto l^{-(\alpha+1)}$, where α is related to β as $\alpha = \beta - 1$ for an exponential sampling of the diameter distribution [27]. By controlling the diameter distribution of the spheres in a Lévy glass we can control α and, thus, the degree of superdiffusion of the material. For $\alpha \geq 2$ the system is diffusive, whereas for $0 < \alpha < 2$ the system is superdiffusive. However, in real systems the finite size of the sample truncates the step-length distribution to the largest sphere diameter, enriching the physics behind the transport properties [28–30]. In previous publications [14, 27] Lévy glasses were fabricated between two microscope slides. In contrast, in this work we remove one of the two slides to reduce undesirable reflections which affect the quality of the measurements. Moreover, the thickness of the sample is approximately 70 µm more than the largest sphere, which means that the step length distribution is truncated on a length scale that is slightly below the sample thickness.

The setup employed follows a standard scheme for coherent backscattering experiments. Light emitted by a HeNe laser (@632 nm) is expanded to collimated beam of 1 cm in diameter to ensure a high angular resolution of the system. A beamsplitter is used to separate the backscattered light from light impinging on the sample. Subsequently, the backscattering cone is imaged on a CCD camera and the use of a polarizer ensures that we observe only the polarization conserving channel [17]. The angular resolution of the setup has been optimized to properly investigate the different shapes of the measured cones. During the data acquisition the sample is nutated to average over different realizations of disorder.

In order to verify the quality of the setup we fabricate a set of diffusive samples made of TiO_2 and polymer (without glass spheres) and measured its backscattering cone. The amount of polymer was adjusted to have the same average (over the total volume) density of scatterers in both diffusive and superdiffusive samples. A cross-cut of the measured diffusive cones is shown in Fig. 2(a) to-

gether with a fit (red curve) obtained from the diffusion theory [21]. The retrieved transport mean free path is $\ell^\star \simeq 19$ µm. In the inset the residuals are displayed as blue dots. The good match between experimental data and fit shows the quality of the experimental setup.

We measured the backscattering cone on two different sets of superdiffusive samples characterized by $\alpha = 1.5$ and $\alpha = 1$. The experimental results are shown in Fig. 2(b) and (c). By comparing these figures one can notice the rising of the tail of the cone as the degree of superdiffusion increases, i.e. when α decreases, following a trend which has been theoretically predicted in Ref. [26]. To ensure that the Lévy cones are not merely *broader* diffusive cones we made a best fit using diffusion theory. The result is shown in Fig. 2(b) and (c) as red curves and the residuals of such fits are shown in the inset of Fig. 2(a). It is clear that the diffusion theory cannot properly describe the optical properties of Lévy glasses.

The coherent backscattering cone for the superdiffusive samples under investigation can be calculated by taking advantage of the fractional derivative approach developed in Ref. [26]. The transport of light in a finite, translationally invariant, superdiffusive system for a point source at x_0 is described by the stationary fractional diffusion equation [26]:

$$D_\alpha \left(\nabla_x^\alpha - k_\perp^\alpha \right) f(x, x_0, \mathbf{k}_\perp) = -\delta(x - x_0), \qquad (1)$$

where ∇^α is the symmetric Riesz fractional derivative with respect to spatial derivatives, $\mathbf{k}_\perp$ is the in-plane component of the wavevector in free space, $f(x, x_0, \mathbf{k}_\perp)$ is the intensity propagator and D_α is a generalized diffusion constant. The spatial nonlocality of ∇^α makes the definition of boundary conditions non trivial [31]. In order to model physical systems, such as Lévy glasses, the fractional Laplacian operator can be represented by an $M \times M$ matrix, whose eigenvalues λ_i, rescaled as $\lambda_i \to \lambda_i(M/L)^\alpha$ with L the slab thickness, and eigenvectors ψ_i converge to those of the continuum operator as M goes to infinity [32]. Absorbing boundary conditions can then be implemented by reducing the infinite size matrix to a finite size matrix and Eq. 1 be solved by eigenfunction expansion. The knowledge of the intensity propagator of the system then makes it possible to calculate interference effects in a "superdiffusion" approximation. In particular, considering a planewave at normal incidence on the slab interface and in the Fraunhofer regime, the coherent component of the albedo is given by the following expression:

$$A_c(\theta) \propto - \sum_{x_1, x_2} F(x_1, x_2, \theta) \sum_{i=1}^{M} \frac{\psi_i(x_1)\psi_i(x_2)}{(\lambda_i - k_\perp^\alpha)}, \qquad (2)$$

where $F(x_1, x_2, \theta) = P(x_1)P(x_2)P(x_1/\cos\theta)P(x_2/\cos\theta)$ describes the attenuation for the amplitude of the incident and emergent planewaves in the scattering medium,
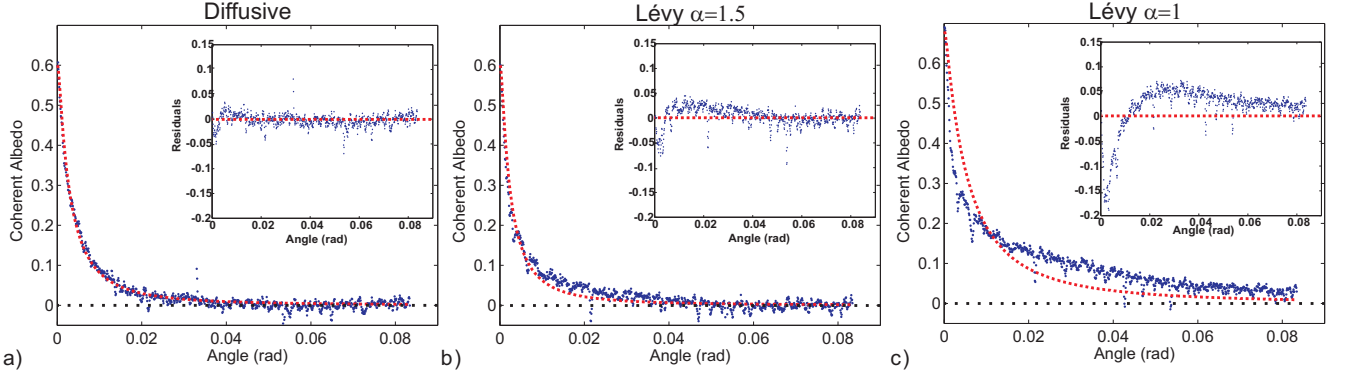
FIG. 2: (a), (b) and (c) Measured coherent backscattering cone from diffusive samples and Lévy glasses with $\alpha = 1.5$ and $\alpha = 1$, respectively. In red the fit to the experimental data according to the standard diffusion theory. In the insets, residuals of the fits.

θ is the angle between the incident and emergent planewaves (see Fig. 1(b)) and $k_\perp = |\mathbf{k}_\perp| \simeq (2\pi/\lambda)\,\theta$, at small angle θ. The amplitude attenuation $P(l)$ was modelled as a Pareto-like distribution $P(l) = 1$ for $0 \le l \le l_c$ and $P(l) = (l_c/l)^{(\alpha+1)/2}$ for $l \ge l_c$, where l_c is the cut-off length, as to closely follow the step length distribution of real Lévy glasses [28]. Internal reflections were neglected.

The results are shown in Fig. 3 for $\alpha = 1.5$ and $\alpha = 1$, where the *only* adjustable parameter used is l_c. The inset shows the residuals between theory and experiment, which are greatly reduced with respect to the diffusive fit (Fig. 2(b) and (c)). Due to the very long tail of the cones for Lévy glasses and the lack of an analytical expression for it, the experimental incoherent background was set to the one obtained semi-analytically. It must be pointed out that for the calculations we employed a step-length distribution which was not truncated, in contrast with the real system. This is due to the fact that the propagator $f(x, x_0, \mathbf{k}_\perp)$ of the system has been calculated by considering the sample sa translational invariant in the in-plane direction and finite in the longitudinal direction. The very good agreement between experiment and calculation shows that the fractional diffusion approach can properly describe light interference effects due to multiple scattering in the superdiffusion approximation.

The gentle slope of the cone tails arising from a Lévy glass is a hallmark of anomalous transport of light. This is a striking effect since generally the width of the cone is expected to be inversely proportional to the average step length ℓ, whereas in a Lévy glass the average step length ℓ_α increases when α decreases. The knowledge of the peculiar Green's function inside this medium, which dictates the shape of the cone, might shed light on this phenomenon. The behavior of such a propagator as a function of the degree of superdiffusion can be retrieved by a Fourier analysis of the experimentally observed coherent backscattering. In diffusive media the coherent albedo can be approximated with the propagator of the
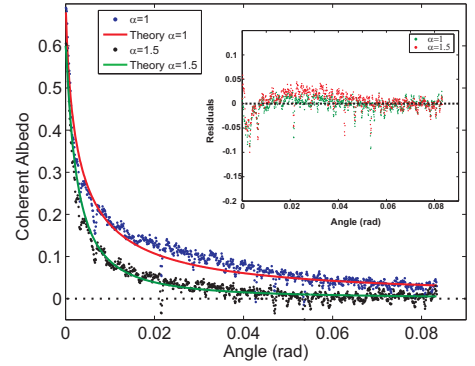

FIG. 3: Comparison between the calculated superdiffusive cones obtained with the fractional derivative approach and the measured Lévy cones.

system in reciprocal space. The propagator is taken for a point source at a distance of a mean free path away from the boundary ($A_c \propto f(x = \ell, x_0 = \ell, k_\perp)$) [33]. We expect a similar Fourier analysis on the coherent albedo from Lévy glasses to provide information on the intensity distribution inside a superdiffusive media. In Fig. 4(a) the normalized Fourier transform of the measured and calculated coherent albedo as a function of the in-plane displacement $\rho = |(\mathbf{r}_i - \mathbf{r}_e)_\perp|$ are shown. These curves are found to be in good agreement, in particular at relatively small values of ρ, and show a remarkable reshaping as a function of α. In Fig 4(b) and (c) the normalized Fourier transform of the theoretical cone and the normalized intensity distribution $f(x = \ell_\alpha, x_0 = \ell_\alpha, \rho)$ calculated from the fractional diffusion approach, respectively, are shown for long ρ. The qualitative agreement between these two figures is evident, in particular in their dependency on the degree of superdiffusion. The spatial distribution of the propagator is dictated by the power-law step length distribution in Lévy glasses, which allows light to couple directly to the surrounding environment from *any* depth inside the sample. This spatial non-locality ap-
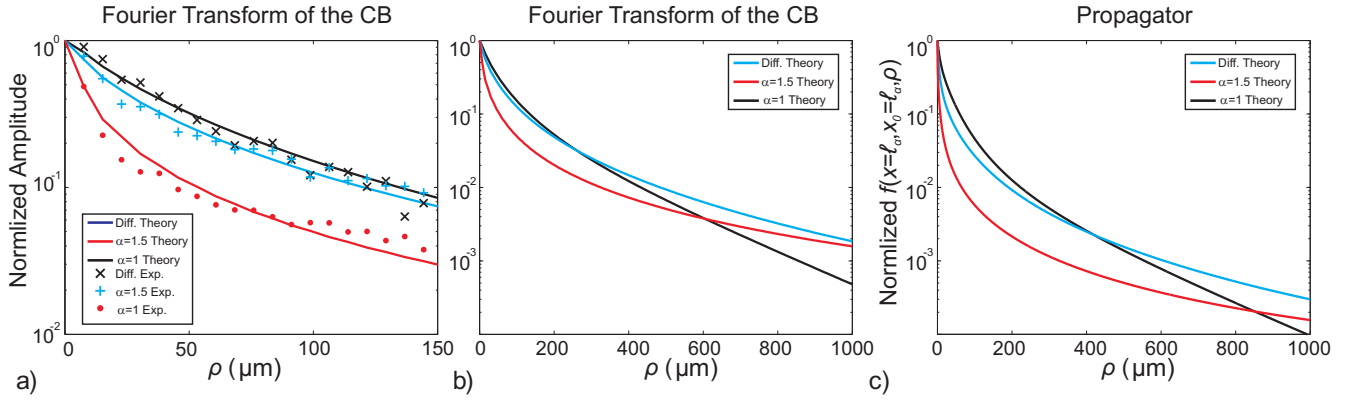
FIG. 4: (a) Amplitude of the Fourier transform of the measured and calculated coherent backscattering (CB) for small ρ. (b) and (c) Amplitude of the Fourier transform of the calculated cone and calculated intensity distribution $f(x = \ell_\alpha, x_0 = \ell_\alpha, \rho)$ for large ρ.

plied to an open system such as a Lévy glass results in a strong modification of the shape of the propagator as a function of α [26]. Firstly, the more cusped shape of the $f(x = \ell_\alpha, x_0 = \ell_\alpha, \rho)$ leads to a smaller in-plane displacement and thus to higher tails in the coherent albedo. Secondly, the gentle tail of the propagator, which induces a cusped cone [26], is a consequence of the fact that light can escape more easily from its local environment towards large distances.

In conclusion, we have reported on the experimental observation of weak localization from Lévy glasses and found strong deviations from the prediction of the diffusion theory. The recently developed semi-analytical model for superdiffusive system is able to reproduce the experimental results with a high degree of accuracy. From the study of the coherent backscattering cone we can retrieve the behavior of the Green's function inside real systems in which the long steps probability is non-vanishing, not only in Lévy glasses but possibly also in foams, clouds or strongly heterogeneous porous media. Further coherent backscattering studies oriented to investigate the influence of real systems parameters (e.g., finite size, truncation, quenching) on the superdiffusive backscattering and their fingerprints in the dynamics of the backscattering cone can be performed to unravel the optical properties of real superdiffusive systems.

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